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WITH HIGH POWER AND BROAD TUNING RANGE

by

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CORRELATED TUNING TRI-FREQUENCY DYE LASER
WITH HIGH POWER AND BROAD TUNING RANGE

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Abstract: A tri-frequency dye laser with coaxial and synchronous output was developed; it can be tuned in the range of 554-667 nm when an R560/DCM alcohol solution is used. It is easy to adjust the power ratio of three wavelengths to be 1:1:1 in the range of 560-640nm based on the correlated tuning principle. An amplifier operation with high power was also achieved.

1. Introduction

It is well known that a dye laser can be used to continuously tune laser wavelengths because the dye can fluoresce over a broad wavelength range. Normally, the radiation spectrum range and radiation power of each dye laser are associated with many factors, including the pump, solvent, and solution concentration. Generally speaking, the tuning range of a dye laser is only from more than a dozen to dozens of nm, and within this range the laser power can vary considerably. In this case, to realize laser tuning with high conversion efficiency in a broader spectrum range, it is necessary to apply multiple dye solutions as alternatives.

In many applications, such as multi-photon spectra, laser isotope separation, laser medical therapy as well as military fields, including the target and background-oriented optical properties research, battlefield smoke spectrum transmittance measurement, laser multi-wavelength remote sensing and reconnaissance, etc., a laser is required not only to accomplish continuous tuning over the broadest possible range, but also to achieve coaxial and synchronous output of several tunable laser wavelengths with similar intensity or adjustable intensity ratio. To reach this goal, it is needed to broaden the radiation spectrum range of dye solutions, and to study and solve the mutual suppression of various wavelength intensities, caused by the competition mechanism of common gain volume patterns during multi-wavelength tuning. Otherwise, it would be difficult to achieve the continuous tuning operation over a broad range at multiple wavelengths, and moreover the actual application requirements could hardly be met due to a great difference in the intensity of the output wavelength.

In the past few years, under the sponsorship of the National Natural Sciences Foundation, we studied the problem of "oscillation and amplification of a correlated tuning multi-frequency dye laser", and achieved remarkable progress and valuable results of practical interest. Additionally, we derived several mixed dye solutions capable of emitting over a broad range[1], and realized, based on the correlated tuning principle, the coaxial correlated tuning amplification operation with double-frequency single dye and mixed dye lasers[2,3]. This paper introduces a tri-frequency dye laser system which is able to accomplish common gain volume tuning. By using a R560/DCM alcohol solution, a single peak continuous tuning curve was obtained through pumping with a 532nm pulse laser; its wavelength range was 554-667nm, and coaxial amplification output was realized with a power ratio of three wavelengths 1:1:1.

2. Spectrum and Laser Tuning Properties of R560 and DCM Alcohol Solutions

The R560 and DCM alcohol solutions pumped with a 532nm laser had a concentration $1-3x10^{-3}$mol. With an absorption peak wavelength 530nm that matches the pumping light, the R560 solution can accomplish high efficiency spectrum conversion; its radiation exhibited ranges from 540 to 610nm with a peak value located near 560nm. While the DCM solution has an absorption peak wavelength near 480nm, yet its absorption band has a broad extension in the long wave region, and therefore, it is also suitable for the 532nm optical pump; also its radiation range obtained is 590-680nm and its peak wavelength varies greatly with concentration, approximately 625±15nm.

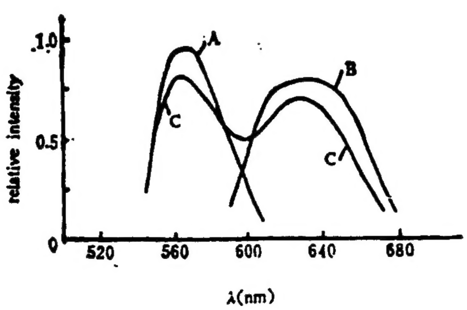

Fig. 1. Emission spectra of dyes R560 and DCM

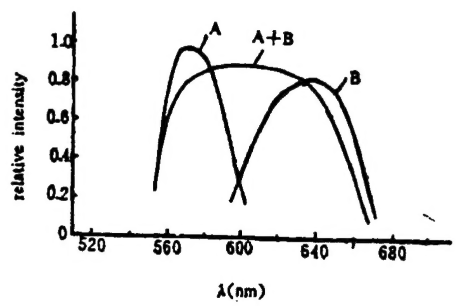

Fig. 2. Laser tuning curves of R560 and DCM dye solutions

The curve A(R560) and curve B(DCM) as shown in Fig. 1 are the radiation spectrum intensity distributions of these two dyes when their concentration was $2x10^{-3}$mol. By mixing the two dyes according to the concentration ratio R560: $2x10^{-3}$mol and DCM: 1 to $2x10^{-3}$mol, the fluorescence intensity distribution was acquired, which is indicated by curve C in the Figure. Curve C suggests that the two dyes can not only absorb pump light simultaneously and luminesce independently, but also execute energy transfer among molecules. The radiation spectrum, in fact, is a integrated contribution of the molecules of the two dyes. With the increase in the total number of dye molecules

involved in radiation, the area ratio below curve C is a little smaller than the sum of the areas below curves A and B, yet much larger than the area below either A or B, indicating that this mixed system possesses ideal spectrum properties, including high spectrum conversion efficiency and broad radiation range.

When this mixed system is used in laser tuning, fairly ideal results are derived as shown in Fig. 2, i.e., the wavelength range of the tuning curve A+B is nearly the sum of the wavelength ranges of the two single dye tuning curves A (R560) and B(DCM).

It is to be noted that the curve A+B is single-peak and continuous, whose peak is located at the junction of the radiation spectra of the two single dyes, and has a fairly flat broad wavelength range; its peak efficiency is equivalent to around 90% of the single dye R560 but higher than the single dye DCM. It was confirmed experimentally that this tuning property is attributed to the resonant energy transfer process occurring in this mixed system, and, in particular, the laser oscillation is a joint gain contribution of the two dye molecules at the junction of the radiation spectra of the two single dyes. This will help us to realize tri-wavelength laser continuous tuning operation in this broad spectrum range.

3. Design of Tri-wavelength Laser Oscillator

Each of the tunable laser wavelengths, respectively, should be generated due to the oscillation in its own resonant cavity. When the three resonant cavities share one common gain volume to produce oscillation with different laser wavelengths, due to the existence of the gain competitive effect, the intensity of the small gain laser wavelength is suppressed by the large gain laser wavelength; as a result, only two or one laser wavelength can survive under critical conditions, while the other one or two wavelengths will not generate optical oscillation. This is a

major problem to be solved in designing a common gain volume tri-wavelength laser oscillator.

The conditions for stable common gain volume tri-wavelength laser oscillation are as follows:

$$\begin{cases} \varepsilon_1 > 1 \\ \varepsilon_1 < 2m_1 \Big/ \Big[(m_1 + 1/2) \Big(\dfrac{\varepsilon_3 + \varepsilon_2}{\varepsilon_2 \varepsilon_3} - 1 \Big) \Big] \\ (\varepsilon_3 + \varepsilon_2) / (\varepsilon_2 \varepsilon_3) < m_1 + 1/2 \end{cases} \tag{1}$$

$$\begin{cases} \varepsilon_2 > 1 \\ \varepsilon_2 < 2m_2 \Big/ \Big[(m_2 + 1/2) \Big(\dfrac{\varepsilon_3 + \varepsilon_1}{\varepsilon_1 \varepsilon_3} \Big) - 1 \Big] \\ (\varepsilon_3 + \varepsilon_1) / (\varepsilon_1 \varepsilon_3) < m_2 + 1/2 \end{cases} \tag{2}$$

$$\begin{cases} \varepsilon_3 > 1 \\ \varepsilon_3 < 2m_3 \Big/ \Big[(m_3 + 1/2) \Big(\dfrac{\varepsilon_2 + \varepsilon_1}{\varepsilon_1 \varepsilon_2} \Big) - 1 \\ (\varepsilon_2 + \varepsilon_1) / (\varepsilon_1 \varepsilon_2) < m_3 + 1/2 \end{cases} \tag{3}$$

where $(\varepsilon_i = g_i/\zeta_i$ $(i=1,2,3)$ is the ratio between gain and loss of different laser wavelengths (briefly, the gain/loss ratio); m_i is the number of longitudinal modes contained in different wavelengths. It is understood, then, that in order for the three wavelengths to produce laser oscillation simultaneously, the gain/loss ratio of different wavelengths must be greater than 1, and also, the gain/loss ratio of the other two wavelengths must be limited in a particular range. In other words, only when the foregoing nine formulas are satisfied simultaneously, can the stable laser oscillation operation of the three wavelengths be realized simultaneously.

Thus, designers must first make sure that the inherent loss of three common gain volume resonant cavities is roughly

consistence, i.e., make the laser tuning performance of the three resonant sub-cavities basically symmetric. Furthermore, designers must insert a continuously variable supplementary loss in the cavity capable of generating a higher gain wavelength, which is used to adjust and control the tuning wavelength gain/loss ratio of different sub-cavities. This is designed to broaden the tri-wavelength laser continuous tuning range, and at the same time, to change the intensity ratio of the output laser wavelength so that the application requirements can be met.

Such a tuning concept or technique, which is based on the gain competition mechanism and loss correlation compensation, is call the correlated tuning principle. The tri-wavelength laser oscillator in a common gain volume, designed on this principle is shown in Fig. 3.

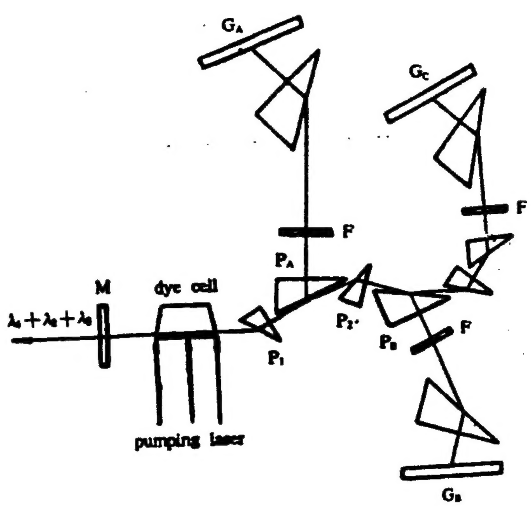

Fig. 3. Tri-wavelength laser oscillator in a common gain volume

Fig. 3 shows a compound Matson resonant cavity structure, where P_A and P_B are beam splitting prisms, which can split a beam emitted from a dye pool into three beams through reflection, refraction and coupling; G_A, G_B and G_C are plates, which,

respectively, form tuning oscillation sub-cavities in common gain volume together with the output coupling reflector. Apart from the beam-splitting function, P_A and P_B, like other prisms, can also expand beams. To make the inherent loss of the three sub-cavities approximately identical with a largest possible energy feedback rate, we employed a simple symmetry design computing technique[5]. F is a rotary Fresnel glass plate inserted in the beam-expanding path, which can be used to broaden the tri-wavelength laser continuous tuning range, and to adjust the intensity ratio of the output laser wavelengths, eventually realizing the co-called correlated tuning.

This design has the following features: 1. Prisms P_A and P_B are incident at the Brewster angle, while other prisms and rasters also have high selectivity over the polarized light vibrating parallel to the paper surface (4 sp component), coupled with the similar selectivity of the active dye molecule diffusion and re-orientation effect[6]. As a result, the tuning lasers generated from different sub-cavities, respectively, have the same polarization direction and a high polarization degree.
2. The number of prisms used for beam expansion in different sub-cavities is optimally selected, with which not only can the requirement for beam expansion times rate be met, but also the reflection loss at the surface of the prisms (one of the fundamental losses of cavities) can be reduced. With this kind of design, i.e., using prisms to split the beam through direct reflection, refraction,and coupling, and the optimal selection of the number of prisms under a given beam expansion times rate, the total energy feedback rate in the cavity is much higher than that in a single sub-cavity. This is favorable not only for extracting the light gain in the gain volume so as to derive intensive oscillation output, but also for broadening the tri-wavelength laser correlated tuning range. 3. The three sub-cavities designed on the principle of symmetry can generate basically the same width of laser lines, and ensure a basically

synchronous output time. 4. The Fresnel glass plate intended for loss correlated compensation tuning is simple and effective, and will not affect the coaxiality of the tri-wavelength laser output while rotating.

4. Experimental Result and Analysis

(1) Selection of Concentration of Dye Solutions

R560 can be directly made into a highly concentrated mother solution using alcohol solvent, while DCM needs to be first made into a highly concentrated mother solution with DMSO solvent before being diluted with alcohol. The attached table lists the laser tuning results of these two single dye solutions with different concentrations. The pump used in the dye pool is 10mm wide with a pump light energy 2-3mJ. It can be seen from the table that the laser wavelength of the two dyes will produce blue shift or red shift at either a low concentration or a high concentration. Further, when the concentration is overly high or too low, both the laser tuning range and peak intensity are small. The peak wavelength of DCM varies to a greater degree with concentration. Only with an appropriate concentration can a higher peak intensity and a broader tuning range be derived. The proper concentrations of the two single dye solutions, respectively, are: 2×10^{-3}mol for R560, and $1-2\times10^{-3}$mol for DCM.

TABLE. Experimental Tuning Values of Dye Solutions with Different Concentrations

R560				DCN		
peak relative intensity	peak wavelength (nm)	wavelength range (nm)	concentration ($\times 10^{-3}$mol)	wavelength range (nm)	peak wavelength (nm)	peak relative intensity
			0.25	595~650	910	21
			0.50	590~660	620	42
40	554	545~584	1.00	591~671	630	46
45	560	554~601	2.00	594~667	635	39
53	560	556~603	3.00	596~662	640	35
44	562	558~600	4.00	605~659	640	18

In selecting the concentration of the mixed dye solution, not only should the pump light absorption conversion rate of single dye solution molecules be taken into account, but also we should also consider the impact of the energy transfer effect between dye molecules on the radiation property, as well as the overall concentration-related quenching effect of the mixed solution. Only in this way can a broad and continuous laser tuning range, and a high efficiency pump light absorption conversion rate of the solution be achieved.

We selected the mixed solution concentration ratio at the oscillation level as R560/DCM=2×10^{-3}mol/1.2×10^{-3}mol, with which we achieved a laser tuning range 554-667nm, its tuning property being shown in the curve A+B in Fig. 2.

The concentration ratio at amplification level should be determined based on factors that include pump power, dye pool width, and the signal light intensity output at the oscillation level. At a pump power approximately 20mJ and a horizontal pumping dye pool width 20mm, we selected the concentration half the value at the oscillation level with which obtained first level amplification output energy greater than 3.5mJ.

(2) Mixed Dye Tri-wavelength Laser Tuning

The R560/DCM mixed alcohol solution was placed in a dye pool as shown in Fig. 3, and one sub-cavity was cut off (i.e., it was prevented from generating laser oscillations). First, the double-wavelength laser tuning experiment was conducted using two sub-cavities as follows: the laser wavelength of one sub-cavity was tuned to approximately 590nm, which was located at the junction of the radiation spectra of two dyes; the other cavity was tuned, respectively, toward the short waves and long waves. It was found that when it was tuned in the range 560-640nm, the intensity of the 590nm laser wavelength did not change greatly, indicating that the double-wavelength laser tuning of the mixed system was much better than that of a single dye. During the double-wavelength laser tuning using either R560 or DCM, when one sub-cavity was tuned to the peak wavelength (approximately 560nm and 630nm, respectively), the other sub-cavity cannot lase even at 590nm. This indicates that the laser oscillations of this mixed system is virtually a result of integrating gain contribution brought about by the independent luminescence and the simultaneous energy transfer mechanism of the two dye molecules. This system can not only achieve a double-wavelength laser tuning range which is much broader than that with a single dye, but also a roughly equivalent double-wavelength laser intensity which executes tuning within this range, i.e., the two laser wavelengths could achieve a laser oscillation output simultaneously even without correlated compensation tuning. This indicates that the coherence between the double-wavelength lasers produced by this system is weaker than in the case of single dyes.

Then, a tri-wavelength laser tuning experiment was conducted in which the laser wavelength of one sub-cavity was first tuned to 590nm, while the other two sub-cavities, respectively, were tuned from 590nm to short wave and long wave directions. It was found that when the tuning was in the range 560nm to 640nm, all

of the coaxial output lasers contain three wavelengths. We tuned two sub-cavities to, respectively, 560nm and 640nm, and tuned the other sub-cavity at least 590±25nm; as a result, we also achieved tri-wavelength laser simultaneous and coaxial oscillation output. Moreover, the tri-wavelength laser oscillation output was also realized when the other one sub-cavity was tuned at a little less than 560nm and a little more than 640nm, indicating that this mixed system as well as the designed tri-wavelength laser oscillator are capable of realizing an extremely broad tri-wavelength laser continuous tuning range even without the correlated compensation tuning.

When the correlated compensation tuning was made on the sub-cavity that generated the intensive laser wavelength through supplementary loss at the Fresnel glass plate, the coaxial output tri-wavelength laser could achieve a continuous tuning in the range from 555nm to 666nm. The experiment demonstrated that with this correlated compensation tuning approach, and when the other wavelength is a little shorter than 560nm and a little longer than 640nm, it is easy to control the tri-wavelength laser output intensity ratio as 1:1:1 in the range 560nm-640nm.

5. Conclusions

The R560/DCM mixed alcohol dye solution is a laser medium with a high efficiency spectrum conversion capability. Due to the pump light absorption of the two dyes' molecules and the presence of resonant energy transfer process, the oscillation laser wavelength can acquire integrating gain contribution. In this case, the laser tuning wavelength range can reach 554nm-667nm, which is nearly the sum of the tuning ranges of the two single dye solutions, and the tuning curve appears single-peak and continuous. Thus, the high power laser amplification operation of this mixed system can be achieved.

Based on the common gain volume multi-wavelength laser correlated tuning principle, and the application of prisms in beam splitting through direct reflection, refraction, and coupling, as well as designing the tri-wavelength dye laser oscillator with the symmetry technique, the coaxial output tri-wavelength laser continuous tuning can be made possible in the range from 555nm to 666nm using R560/DCM mixed alcohol solution and correlated compensation tuning. The tri-wavelength laser shows a higher conversion efficiency at least in the range 560nm-640nm, and the output intensity ratio is easy to be controlled within 1:1:1.

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